

At page 1, please AMEND the PARAGRAPH beginning on line 3 with the words "The United States Government has rights ..." as shown below.

At page 1, please AMEND the PARAGRAPH beginning on line 17 with the words "Hydrides of aluminum with lithium ..." as shown below.

At page 2, please AMEND the PARAGRAPH beginning on line 13 with the words "However, the alkali metal hydrides..." as shown below.

At page 2, please AMEND the PARAGRAPH beginning on line 23 with the words "The present invention provides..." as shown below.

At page 4 please AMEND the PARAGRAPH beginning on line 23 with the words "FIGURES 9..." as shown below.

At page 5 please AMEND the PARAGRAPH beginning on line 1 with the words "The hydrides of alkali metals..." as shown below.

At page 5 and continuing to page 6, please AMEND the PARAGRAPH beginning on line 27 with the words "In a particular embodiment..." as shown below.

At page 8 please AMEND the PARAGRAPH beginning on line 11 with the words "Initial hydrogenation is shown..." as shown below.

At page 8 and continuing to page 9, please AMEND the PARAGRAPH beginning on line 26 with the words "The preparation of the catalyzed..." as shown below.

At page 9, please AMEND the PARAGRAPH beginning on line 10 with the words "After milling..." as shown below.

At page 9, please AMEND the PARAGRAPH beginning on line 19 with the words "In the initial absorption..." as shown below.

At page 10, please AMEND the PARAGRAPH beginning on line 4 with the words "A final example for the fabrication..." as shown below.

At page 10, please AMEND the PARAGRAPH beginning on line 21 with the words "Evidence of the formation..." as shown below.

At page 10, please AMEND the PARAGRAPH beginning on line 26 with the words "The foregoing examples,..." as shown below.

AMENDMENTS TO THE SPECIFICATION

ON PAGE 1

AT ORIGINAL LINE 3 OF THE SPECIFICATION, IN THE PARAGRAPH BEGINNING WITH THE WORDS "The United States Government has rights ..." the operation of the Sandia National Laboratories." PLEASE AMEND THE SPECIFICATION WITH THE REPLACEMENT PARAGRAPH AS FOLLOWS:

STATEMENT OF GOVERNMENT INTEREST

The United States Government has rights in this invention pursuant to Contract No. DE-AC04-94AL85000 between the United States Department of Energy and Sandia Corporation, for the operation of the Sandia National Laboratories.

ON PAGE 1

AT ORIGINAL LINE 17 OF THE SPECIFICATION, IN THE PARAGRAPH BEGINNING WITH THE WORDS "Hydrides of aluminum with lithium ..." AND ENDING WITH THE WORDS "...production of LiAlH_4 , NaAlH_4 , KAlH_4 and CsAlH_4 ." PLEASE AMEND THE SPECIFICATION WITH THE REPLACEMENT PARAGRAPH AS FOLLOWS:

Hydrides of aluminum with lithium, sodium, and potassium have been ~~know~~ known for many years. A direct synthesis method to produce these materials was first described (French Patent Serial Number 1,235,680). According to Ashby, synthesis of, for instance, NaAlH_4 can be performed by placing either the alkali metal or its hydride into an autoclave with activated aluminum powder in a solvent such as tetrahydrofuran. The mixture is subjected to hydrogen at a pressure of 2000 psi (about 135 atm) and heated to 150°C for several hours after which the mixture is cooled, the excess aluminum is separated by filtration, and the NaAlH_4 isolated by precipitation using a hydrocarbon additive such as toluene to the tetrahydrofuran solution, followed by vacuum distillation of the tetrahydrofuran. The method is applicable to the production of LiAlH_4 , NaAlH_4 , KAlH_4 and CsAlH_4 .

ON PAGE 2

AT ORIGINAL LINE 13 OF THE SPECIFICATION, IN THE PARAGRAPH BEGINNING WITH THE WORDS "However, the alkali metal hydrides ..." AND ENDING WITH THE WORDS "...reported by Bogdanovic, et al." PLEASE AMEND THE SPECIFICATION WITH THE REPLACEMENT PARAGRAPH AS FOLLOWS:

However, the alkali metal hydrides prepared in the traditional manner act only to irreversibly release hydrogen under moderate conditions. While Bogdanovic, et al., (U.S. Patent Number 6,106,801) have reported that ~~that~~ the addition of a transition metal compound ~~act~~ acts as a catalyst to aid in the re-absorption of hydrogen, the kinetics of this system have been reported to be slow and unstable. Zaluska, et al., (U.S. Patent Number 6,251,349) have reported reversible absorption and desorption of hydrogen is achieved in complex alkali metal-aluminum hydride compounds prepared by mechanical mixing/milling mixtures of the simple hydrides without the catalyst reported by Bogdanovic, et al.

ON PAGE 2

AT ORIGINAL LINE 23 OF THE SPECIFICATION, IN THE PARAGRAPH BEGINNING WITH THE WORDS "The present invention provides ..." AND ENDING WITH THE WORDS "...reversible hydrogenation properties." PLEASE AMEND THE SPECIFICATION WITH THE REPLACEMENT PARAGRAPH AS FOLLOWS:

The present invention provides a totally different method for preparing alkali metal-aluminum hydrides which is based on simple a two step-process. The resulting hydrides exhibit outstanding reversible hydrogenation properties.

ON PAGE 4

AT ORIGINAL LINE 23 OF THE SPECIFICATION, IN THE PARAGRAPH BEGINNING WITH THE WORDS "FIGURES 9..." AND ENDING WITH THE WORDS "...to prepare $\text{NaAlH}_4 + \text{Na}_3\text{AlH}_6$." PLEASE AMEND THE SPECIFICATION WITH THE REPLACEMENT PARAGRAPH AS FOLLOWS:

FIGURE 9 ~~show~~ shows Arrhenius plots of the rates of hydrogen desorption from $\text{NaH} + \text{Al}$ samples that were hydrided after doping with three different levels of TiCl_3 or TiF_3

using the direct synthesis method of the present invention to prepare NaAlH_4 + Na_3AlH_6 .

ON PAGE 5

AT ORIGINAL LINE 1 OF THE SPECIFICATION, IN THE PARAGRAPH BEGINNING WITH THE WORDS "The hydrides of alkali metals ..." AND ENDING WITH THE WORDS "...and methods of preparation." PLEASE AMEND THE SPECIFICATION WITH THE REPLACEMENT PARAGRAPH AS FOLLOWS:

The hydrides of alkali metals and aluminum are compounds that belong to the larger class of complex hydrides. These compounds are known to liberate copious amounts of hydrogen either by direct thermal decomposition or by one-time hydrolysis. However, they were generally considered too irreversible for practical hydrogen storage applications. This was until Bogdanovic, et al., (Bogdanovic and Schwickardi, J. Alloys and Compounds, vol. 253, no. 1, 1997) demonstrated that NaAlH_4 , would reversibly desorb and ~~absorbed~~ absorb hydrogen under relatively mild conditions when doped with one of a number of catalyst compounds. Since that time there has been a growing body of work in characterizing catalyzed alkali metal-aluminum hydrides, as well as the development of new catalysts and methods of preparation.

ON PAGE 5 AND CONTINUING TO PAGE 6

AT ORIGINAL LINE 27 OF THE SPECIFICATION, IN THE PARAGRAPH BEGINNING WITH THE WORDS "In a particular embodiment, ..." AND ENDING WITH THE WORDS "...or near room temperature." PLEASE AMEND THE SPECIFICATION WITH THE REPLACEMENT PARAGRAPH AS FOLLOWS:

In a particular embodiment, fabrication of alkali metal-aluminum hydrides comprise mixing powders of a simple alkali metal hydride (LiH , NaH , KH) with aluminum powder and a transition metal catalyst compound (typically a titanium catalyst compound such as TiCl_3 , TiF_3 , or mixtures of equal parts of these materials) in the desired proportion and ball milling the constituents under an inert atmosphere of argon gas. (While not attempted, other dry ~~gas~~ gases such as helium, hydrogen are also believed to be effective). The milling step is carried out at or near room temperature.

ON PAGE 8

AT ORIGINAL LINE 11 OF THE SPECIFICATION, IN THE PARAGRAPH BEGINNING WITH THE WORDS "Initial hydrogenation is shown ..." AND ENDING WITH THE WORDS "...within about 2.2 hours." PLEASE AMEND THE SPECIFICATION WITH THE REPLACEMENT PARAGRAPH AS FOLLOWS:

Initial hydrogenation is shown in FIGURE 3 as the first half-cycle (1st absorption) in this example. Again, the sample reaches about 90% of ~~it~~ its maximum capacity within about 20 hours. Subsequent absorption/desorption half-cycles (2nd and 3rd absorption and 1st and 2nd desorption) for this example are shown in FIGURES 3 and 4. The rates of each subsequent absorption half-cycle after the initial half-cycle are seen to improve from under about an hour for 80% of maximum capacity, and to about 5 hours for 90% of capacity. FIGURE 4 shows desorption of hydrogen to about 60% of capacity was achieved within about 1 hour and to about 90% of capacity within about 2.2 hours.

ON PAGE 8 AND CONTINUING TO PAGE 9

AT ORIGINAL LINE 26 OF THE SPECIFICATION, IN THE PARAGRAPH BEGINNING WITH THE WORDS "The preparation of the catalyzed ..." AND ENDING WITH THE WORDS "...high purity argon gas atmosphere." PLEASE AMEND THE SPECIFICATION WITH THE REPLACEMENT PARAGRAPH AS FOLLOWS:

The preparation of the catalyzed hydrides $\text{NaAlH}_4 + \text{Na}_3\text{AlH}_6$ directly from the alkali metal was demonstrated by mechanical milling 3.03 grams of sodium metal together with 3.56 grams of aluminum metal powder and 0.407 grams of a TiCl_3 catalyst precursor compound (molar ratios of 1.0:1.0:0.02) in a tungsten carbide lined steel vial with tungsten carbide balls in a SPEX™ mill. In this example processing proceeds as before except that the sodium metal is introduced into the ball mill as small slivers or pieces of the cut metal. About one tenth of the quantity is fed into the mill at one time and the contents of the mill are mechanically "worked" for several minutes before more metal is added. This is repeated several ~~time~~ times until the required quantity of sodium is introduced into the mill. Again, the ball mass to sample mass ratio was about

9:1. Total milling time was about 2 to 3 hours and processing took place at near room temperature under a high purity argon gas atmosphere.

ON PAGE 9

AT ORIGINAL LINE 10 OF THE SPECIFICATION, IN THE PARAGRAPH BEGINNING WITH THE WORDS "After milling, ..." AND ENDING WITH THE WORDS "...with a computer." PLEASE AMEND THE SPECIFICATION WITH THE REPLACEMENT PARAGRAPH AS FOLLOWS:

After milling, the material formed a dark, hard, metallic-looking material. About 1.5 grams of the sample was transferred (under argon) to a ~~the~~ stainless steel reactor vessel described in Example 1 and exposed to high purity (99.999 %) hydrogen gas. The hydrogen was again pressurized to between about 80 atm to about 100 atm while the steel reactor and its contents are heated externally with electrical tape to about 125°C for up to 20 hours. Pressure measurements were taken using a calibrated 200 atm pressure transducer for the absorption half-cycle and a 1.3 atm calibrated Baratron™ capacitance manometer for the desorption half-cycle. Data was recorded with a computer.

ON PAGE 9

AT ORIGINAL LINE 19 OF THE SPECIFICATION, IN THE PARAGRAPH BEGINNING WITH THE WORDS "In the initial absorption ..." AND ENDING WITH THE WORDS "...desorption half-cycle." PLEASE AMEND THE SPECIFICATION WITH THE REPLACEMENT PARAGRAPH AS FOLLOWS:

In the initial absorption half-cycle shown in FIGURE 5 (1st absorption) the formation of NaAlH₄ is preceded by the formation of NaH during the first high temperature excursion at a temperature of about 200°C. Subsequent NaAlH₄ formation took place at temperatures between 80°C and 120°C (100°C shown). Greater than 90% of the hydrogen absorption is seen ~~takes~~ taking place within about a 2 hour period during the initial hydrogenation step. The results demonstrate that NaAlH₄ can be prepared from Na metal without the need for a separate process to produce NaH. FIGURE 6 shows the material to be fully reversible, releasing about 2 weight percent hydrogen gas in the desorption half-cycle.

ON PAGE 10

AT ORIGINAL LINE 4 OF THE SPECIFICATION, IN THE PARAGRAPH BEGINNING WITH THE WORDS "A final example for the fabrication ..." AND ENDING WITH THE WORDS "...NaCl during processing." PLEASE AMEND THE SPECIFICATION WITH THE REPLACEMENT PARAGRAPH AS FOLLOWS:

A final example for the fabrication of $\text{NaAlH}_4 + \text{Na}_3\text{AlH}_6$ was performed as in EXAMPLE 1 to test the effects of a range of catalyst content on the hydrogenation behavior of the hydride. In this example 3.13 grams of NaH was ball milled together with 3.145 grams of aluminum metal powder and 0.720 grams of the TiCl_3 catalyst precursor compound (molar ratios of 1.12:1.0:0.04). Again, milling was preformed in a tungsten carbide lined steel vial with several tungsten carbide balls. Again, excess alkali metal hydride is added to account for the formation of NaCl during processing.

ON PAGE 10

AT ORIGINAL LINE 21 OF THE SPECIFICATION, IN THE PARAGRAPH BEGINNING WITH THE WORDS "Evidence of the formation ..." AND ENDING WITH THE WORDS "...shown in FIGURE 10." PLEASE AMEND THE SPECIFICATION WITH THE REPLACEMENT PARAGRAPH AS FOLLOWS:

Evidence of the formation of a quantity of $\text{NaAlH}_4 + \text{Na}_3\text{AlH}_6$ was again confirmed by the x-ray diffraction pattern shown in FIGURE 10. The spectra was obtained (using an airless sample holder) from a portion of the material after the 2nd hydriding cycle and is displayed as the ~~bottom~~ lowest of the three spectra shown in FIGURE 10.

ON PAGE 10

AT ORIGINAL LINE 26 OF THE SPECIFICATION, IN THE PARAGRAPH BEGINNING WITH THE WORDS "The foregoing examples, ..." AND ENDING WITH THE WORDS "...in hydrogen transfer rate." PLEASE AMEND THE SPECIFICATION WITH THE REPLACEMENT PARAGRAPH AS FOLLOWS:

The foregoing examples, therefore, clearly ~~demonstrates~~ demonstrate the formation of sodium aluminum hydride. It is believed that lithium and potassium aluminum hydrides may be prepared using a similar technique. It is demonstrated further, as shown in FIGURE 9, that the use of the titanium catalyst compound, which some have reported to be unnecessary, has a marked and dramatic effect on the rate of hydrogen desorption from the sodium aluminum hydrides compounded by the present method. FIGURE 9 illustrates this effect as multiple Arrhenius plots for several hydride samples that contain various levels of the titanium catalyst compound. These curves clearly show an increase of several orders of magnitude in the kinetics of hydrogen desorption by the doped hydride material over the undoped material and the effect appears to require only a modest amount of the catalyst compound in order to obtain the desired increase in hydrogen transfer rate.